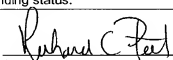


JC10 Rec'd PCT/PTO 19 FEB 2002

FORM PTO-1390 (Modified) (REV 5-93)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER	
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				077251-0104	
INTERNATIONAL APPLICATION NO. PCT/EP00/07950		INTERNATIONAL FILING DATE 08/16/2000		U.S. APPLICATION NO. (if known) 37 C.F.R. 1.55 Unassigned <b>10/049839</b> PRIORITY DATE CLAIMED 08/19/1999	
TITLE OF INVENTION FLOWABLE ALKALINE FLUORIDES AND ALKALINE EARTHS FLUORIDES					
APPLICANT(S) FOR DO/EO/US Andreas KLINGENBERG					
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:					
<p>1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.</p> <p>2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.</p> <p>3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).</p> <p>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19<sup>th</sup> month from the earliest claimed priority date.</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))  <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).  <input checked="" type="checkbox"/> has been transmitted by the International Bureau.  <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US)</p> <p>6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</p> <p>7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))  <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).  <input type="checkbox"/> have been transmitted by the International Bureau.  <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.  <input checked="" type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</p> <p>10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p> <p>11. <input type="checkbox"/> Applicant claims small entity status under 37 CFR 1.27.</p>					
Items 12. to 17. below concern other document(s) or information included:					
<p>12. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</p> <p>13. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</p> <p>14. <input type="checkbox"/> A FIRST preliminary amendment.  <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</p> <p>15. <input type="checkbox"/> A substitute specification.</p> <p>16. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>17. <input checked="" type="checkbox"/> Other items or information: Application Data Sheet and Preliminary Amendment.</p>					

U.S. APPLICATION No. (37 CFR 1.53) <b>Unassigned 107049839</b>		INTERNATIONAL APPLICATION NO PCT/EP00/07950		ATTORNEY'S DOCKET NUMBER 077251-0104	
18. <input checked="" type="checkbox"/> The following fees are submitted:				<b>CALCULATIONS</b>	
Basic National Fee (37 CFR 1.492(a)(1)-(5): Search Report has been prepared by the EPO or JPO.....\$890.00					
International preliminary examination fee paid to USPTO (37 CFR 1.482).....\$710.00					
No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) .....\$740.00					
Neither international preliminary examination fee (37 CFR 1.482) nor International search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$1,040.00					
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) .....\$100.00					
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				<b>\$890.00</b>	
Surcharge of \$130.00 for furnishing the oath or declaration later than 20 Months from the earliest claimed priority date (37 CFR 1.492(e))					
Claims	Number Filed	Included in Basic Fee	Extra Claims	Rate	
Total Claims	7	- 20	= 0	x \$18.00	\$0.00
Independent Claims	1	- 3	= 0	x \$84.00	\$0.00
Multiple dependent claim(s) (if applicable)				\$280.00	\$280.00
<b>TOTAL OF ABOVE CALCULATIONS =</b>				<b>\$1,170.00</b>	
Reduction by 1/2 for filing by small entity, if applicable.				\$0.00	
<b>SUBTOTAL =</b>				<b>\$1,170.00</b>	
Processing fee of \$130.00 for furnishing English translation later the 20 months from the earliest claimed priority date (37 CFR 1.492(f)).				+	
<b>TOTAL NATIONAL FEE =</b>				<b>\$1,170.00</b>	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +				+	
<b>TOTAL FEES ENCLOSED =</b>				<b>\$1,170.00</b>	
				Amount to be: refunded \$	
				charged \$	
a. <input checked="" type="checkbox"/> A check in the amount of \$1,170.00 to cover the above fees is enclosed.					
b. <input type="checkbox"/> Please charge my Deposit Account No. <u>19-0741</u> in the amount of \$0.00 to the above fees. A duplicate copy of this sheet is enclosed.					
c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>19-0741</u> . A duplicate copy of this sheet is enclosed.					
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.					
SEND ALL CORRESPONDENCE TO:					
Foley & Lardner Customer Number: 22428  <b>*22428*</b>  <b>22428</b>			 SIGNATURE NAME RICHARD C. PEET REGISTRATION NUMBER 35,792		
PATENT TRADEMARK OFFICE					

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

**Attorney Docket No. 077251/0104**

In re patent application of

Andreas KLINGENBERG

Serial No. Unassigned

Group Art Unit: Unassigned

Filed: February 19, 2002

Examiner: Unassigned

For: FLOWABLE ALKALINE FLUORIDES AND ALKALINE EARTHS FLUORIDES

**PRELIMINARY AMENDMENT**

Commissioner for Patents  
Box NON-FEE AMENDMENT  
Washington, D.C. 20231

Sir:

Preliminary to examination, please amend the above-identified application as follows:

**IN THE TITLE:**

Please delete the title of the invention and substitute therefore:

FREE FLOWING ALKALI FLUORIDES AND ALKALINE EARTH FLUORIDES

The changes made to the title are shown in the marked-up version of the specification.

**IN THE SPECIFICATION**

Before the first paragraph of page 1, please insert the following:

This application incorporates German patent application 199 39 353.2, filed August 19, 1999, by reference in its entirety.

IN THE CLAIMS:

Please cancel claims 1-8 and add new claims 9-15 as follows.

9. (New) A mixture, comprising an alkali metal fluoride or an alkaline-earth metal fluoride or a mixture of both and 2% by weight or less of pyrogenic silicic acid.
10. (New) The mixture of claim 9, wherein the alkaline-earth metal is potassium fluoride.
11. (New) The mixture of claim 9, characterized in that it contains from 0.2 to 2% by weight of pyrogenic silicic acid, relative to the total weight of the mixture.
12. (New) The mixture of claim 9, characterized in that it contains from 99 to 99.8% by weight of potassium fluoride and from 0.2 to 1% by weight of pyrogenic silicic acid, each relative to the total amount of the mixture.
13. (New) A process for the manufacture of the mixture of claim 1, comprising mixing (A) a dried alkali metal fluoride or a dried alkaline-earth metal fluoride or a mixture of both and (B) 2% by weight or less of pyrogenic silicic acid, relative to the total weight of the mixture.
14. (New) The process of claim 13, wherein the dried alkali metal fluoride or the dried alkaline-earth metal fluoride or the dried mixture of both is obtained by spray drying of a solution, comprising the alkali metal fluoride or the alkaline-earth metal fluoride or the mixture of both.
15. (New) The process of claim 14, wherein the spray drying is carried out at an inlet temperature in the range of from 300 to 600 °C and an outlet temperature of from 130 to 190 °C.

REMARKS

Claim 1-8 have been canceled without prejudice or disclaimer. With entry of this amendment, claims 9-15 are pending. Applicants await an Office Action on the merits.

Respectfully submitted,

Date

February 19, 2002

Richard C. Peet  
Reg. No. 35,792

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Should additional fees be necessary in connection with the filing of this paper, or if a petition for extension of time is required for timely acceptance of same, the Commissioner is hereby authorized to charge Deposit Account No. 19-0741 for any such fees; and applicant(s) hereby petition for any needed extension of time.

10/11 rec'd PCT/PTO 08 JUL 2002  
10/040339  
#3

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Attorney Docket No. 077251/0104

In re patent application of

Andreas KLINGENBERG

Serial No. 10/049,839

Group Art Unit: Unassigned

Filed: February 19, 2002

Examiner: Unassigned

For: FLOWABLE ALKALINE FLUORIDES AND ALKALINE EARTHS FLUORIDES

SECOND PRELIMINARY AMENDMENT

Commissioner for Patents  
Box NON-FEE AMENDMENT  
Washington, D.C. 20231

Sir:

Prior to examination, please amend the above-identified application as follows:

IN THE SPECIFICATION

Please delete the first paragraph on page 1, added by Preliminary Amendment on February 19, 2002, which reads, "This application incorporates German patent application 199 39 353.2, filed August 19, 1999, by reference in its entirety."

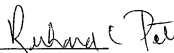
REMARKS

This second preliminary amendment is submitted to delete an incorporation by reference that was mistakenly included in the first preliminary amendment. Applicant awaits an Office Action on the merits.

Respectfully submitted,

July 8, 2002  
Date

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Should additional fees be necessary in connection with the filing of this paper, or if a petition for extension of time is required for timely acceptance of same, the Commissioner is hereby authorized to charge Deposit Account No. 19-0741 for any such fees; and applicant(s) hereby petition for any needed extension of time.

(12) NACH DEM VERTRAG ÜBER DIE INTERNATIONALE ZUSAMMENARBEIT AUF DEM GEBIET DES  
PATENTWESENS (PCT) VERÖFFENTLICHTE INTERNATIONALE ANMELDUNG(19) Weltorganisation für geistiges Eigentum  
Internationales Büro(43) Internationales Veröffentlichungsdatum  
1. März 2001 (01.03.2001)

PCT

(10) Internationale Veröffentlichungsnummer

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C01F 11/22, 5/28, B01J 2/30

(21) Internationales Aktenzeichen: PCT/EP00/07950

(22) Internationales Anmeldedatum:  
16. August 2000 (16.08.2000)

(25) Einreichungssprache: Deutsch

(26) Veröffentlichungssprache: Deutsch

(30) Angaben zur Priorität:  
199 39 353.2 19. August 1999 (19.08.1999) DE(71) Anmelder (für alle Bestimmungsstaaten mit Ausnahme  
von US): HONEYWELL SPECIALTY CHEMICALS  
SEELZE GMBH [DE/DE]; Wunstorfer Strasse 40,  
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(72) Erfinder; und

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(DE).(74) Anwalt: STURM, Christoph; Honeywell Holding AG,  
Kaiserleistrasse 39, D-63067 Offenbach am Main (DE).(81) Bestimmungsstaaten (national): AE, AL, AM, AT, AU,  
AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DK,  
DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL,  
IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU,  
LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT,  
RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA,  
UG, US, UZ, VN, YU, ZA, ZW.(84) Bestimmungsstaaten (regional): ARIPO-Patent (GH,  
GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), eura-  
sisches Patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM),  
europäisches Patent (AT, BE, CH, CY, DE, DK, ES, FI,  
FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI-Patent  
(BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE,  
SN, TD, TG).

## Veröffentlicht:

- Mit internationalem Recherchenbericht.
- Vor Ablauf der für Änderungen der Ansprüche geltenden Frist; Veröffentlichung wird wiederholt, falls Änderungen eintreffen.

Zur Erklärung der Zweibuchstaben-Codes, und der anderen Abkürzungen wird auf die Erklärungen ("Guidance Notes on Codes and Abbreviations") am Anfang jeder regulären Ausgabe der PCT-Gazette verwiesen

(54) Title: FLOWABLE ALKALINE FLUORIDES AND ALKALINE EARTHS FLUORIDES

(54) Bezeichnung: RIESELFÄHIGE ALKALI- UND ERDALKALIFLUORIDE

(57) Abstract: A mixture, comprising an alkaline fluoride or an alkaline earths fluoride or a mixture from two or more thereof and 2 % by weight or less of silicic acid, preferably pyrogenic silicic acid. Also disclosed are the production method and the use of said mixture in the production of high-purity metals, in silver soldering, as a catalyst in the conversion of organic compounds and as a fluorination agent for organic compounds.

(57) Zusammenfassung: Gemisch, umfassend ein Alkalifluorid oder ein Erdalkalifluorid oder eine Mischung aus zwei oder mehr davon und 2 Gew.-% oder weniger Kieselsäure, vorzugsweise pyrogene Kieselsäure, Verfahren zu dessen Herstellung und dessen Verwendung bei der Herstellung von hochreinen Metallen, beim Silberlöten, als Katalysator bei der Umsetzung organischer Verbindungen und als Fluorierungsmittel für organische Verbindungen.

- 1 -

PCT/EP00/07950  
Honeywell Specialty Chemicals Seelze  
GmbH

11. Februar 2002  
R29699PC RI/Ben

Translation of the PCT-application as originally filed

5                   **Flowable alkaline fluorides and alkaline earths fluorides**

The present invention relates to free flowing alkali metal fluorides, earth alkali metal fluorides or mixtures of two or more thereof, a process for the production thereof and the use in the manufacture of highly pure metals, in silver soldering, as catalyst in the reaction of organic compounds and as fluorinating agent for organic compounds.

15           Alkali metal fluorides and earth alkali metal fluorides are important chemicals with versatile industrial applications. For example water free potassium fluoride is used as fluxing agent in the production of highly pure metals, in silver soldering, as catalyst in the organic synthesis and as fluorinating agent for organic compounds.

20           At room temperature water free potassium fluoride has the tendency to form the thermodynamically more stable higher hydrates ( $\text{KF} \cdot 2\text{H}_2\text{O}$ ;  $\text{KF} \cdot 4\text{H}_2\text{O}$ ). Therefor said compound is strongly hygroscopic so that during the use one has to pay attention to the humidity of the environment.

25           Another undesired property of potassium fluoride is its tendency to get hardened. Directly after the manufacture potassium fluoride normally is absolutely free flowing, but gets hardened within a very short time resulting in a constituency comparable to sticky and lumpy snow. During packing in polyethylene internal lining with wrapping or fiber drum potassium fluoride takes on the shape of the cardboard box or the fiber drum and is keeping said form even after removing said

30



- 2 -

jacketing. Therefor the handling of potassium fluoride is extremely difficult because of this property, for example during the filling of a reaction vessel through the manhole. To prevent said baking, for example the baking of potassium fluoride, compounds prior to be ground to fine powders were used till now, as for example talcum, mica, diatomite, hydroxyl apatite, sodium sulfate, stearic acid or sodium polyacrylate, in an amount of some percentage by weight, relative to the mixture of said fluoride and said additive, as it is described in the introductory part of the US 4,806,332. For the desired reduction of the baking tendency the addition of some percentage by weight of said compounds is necessary, so that the resulting alkali metal fluoride or earth alkali metal fluoride, in particular potassium fluoride, is strongly polluted and the requirements of a purity of a content of more than 99 % by weight of alkali metal fluoride and/or earth alkali metal fluoride for some applications cannot be achieved, for example in the production of highly pure metals. Furthermore these additives have to be distributed very homogeneous into the end product to provide full efficiency, what has proved to be impracticable with respect to the large amounts being necessary.

Another suggestion for the prevention of said baking of potassium fluoride is made in the aforementioned application US 4,806,332. Herein a process for the prevention of the hardening of water free potassium fluoride is described, wherein potassium fluoride is brought into contact with gaseous carbon dioxide, adsorbing said gas.

With respect to the prior art as described above it was an object of the invention to provide an alternative process for the production of free flowing, preferably water free alkali metal fluorides and/or earth alkali metal fluorides as well as to provide said compounds per se. The process of the invention should be carried out in a simple manner and should provide a product still being free flowing after a storage period of several months.

This object could be achieved by the process of the invention and by the products resulting from said process.

Accordingly the invention relates to a mixture, comprising an alkali metal fluoride  
5 or earth alkali metal fluoride or a mixture of two or more thereof and 2 % by  
weight or less of silicic acid, preferably pyrogenic silicic acid, as well as a process  
for the manufacture of a mixture, characterized in that dried alkali metal fluoride  
or dried earth alkali metal fluoride or a mixture of two or more thereof is mixed  
with 2 % by weight or less of silicic acid, relative to the total weight of the mix-  
10 ture.

There are no special restrictions concerning the alkali metal fluoride and/or earth  
alkali metal fluoride used in the invention. Preferably the present invention relates  
to mixtures, comprising lithium fluoride, sodium fluoride, potassium fluoride,  
15 magnesium fluoride and/or calcium fluoride, further preferred potassium fluoride.  
It is preferred that the content of alkali metal fluoride and/or earth alkali metal  
fluoride, in particular potassium fluoride, is at least of 99 % by weight, further  
preferred at least of 99.5 % by weight.

20 The mixture of the invention contains further silicic acid, preferably pyrogenic  
silicic acid, further preferred hydrophobized pyrogenic silicic acid, which in par-  
ticular contains an amount of silicon dioxide of more than 99.8 % by weight, rela-  
tive to the total weight of the silicic acid. Said silicic acids are generally highly  
dispersed and provide a very large specific BET-surface of approximately of from  
25 50 to 400 m<sup>2</sup>/g, preferably approximately of from 150 to 400 m<sup>2</sup>/g and in particu-  
lar approximately of from 250 to 400 m<sup>2</sup>/g. Said silicic acid consists of amor-  
phous, preferably ball-shaped particles with a diameter in the range of from 27 to  
40 nm, preferably of from 10 to 20 nm. Said highly dispersed silicic acid, which is  
known as amorphous pyrogenic silicic acid, too, is commercial available under  
30 the trade name Aerosil<sup>®</sup>, wherein among those highly dispersed silicic acids are  
those preferred, which are hydrophobized with silanes, for example dichlorodi-

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methysilane. In particular are to be named: Aerosil®130, Aerosil®150, Aerosil®200, Aerosil®300, Aerosil®380, Aerosil®OX50, Aerosil®TT600, Aerosil®MOX80, Aerosil®MOX170, Aerosil®COK84, and Aerosil®R972, wherein among these Aerosil®R972 is preferred. Furthermore the commercial available  
5 silicic acids which are sold under the trade name Sipernat® of company Degussa and Cab-O-Sil® can be applied.

In the mixture of the invention the silicic acid content is according to the invention up to 2 % by weight, preferably up to 1% by weight, further preferred up to  
10 0.5 % by weight, each relative to the total weight of the mixture.

An especially preferred mixture of the invention contains of from 99 to 99.8 % by weight of potassium fluoride and of from 0.2 to 1 % by weight of silicic acid as defined above.

15

For several of the striven further applications it is desirable by the invention that the mixture is practically water free, i. e. showing a water content after the spray drying of less than 0.3 % by weight, preferably less than 0.2 % by weight and in particular less than 0.1 % by weight, each relative to the total weight of the mixture.  
20

In the process of the invention for the manufacture of the mixture of the invention as described at the beginning the alkali metal fluoride and/or the earth alkali metal fluoride are used in dried form, wherein there are no special restrictions concerning the drying methods for said fluorides. In particular there are to name: vacuum  
25 drying, circulating air drying, fluidized-bed drying, and spray drying, wherein it is preferred to use the spray drying of a solution, comprising said alkali metal fluoride and/or earth alkali metal fluoride. Said spray drying is carried out according to the well known methods of the prior art, wherein the inlet temperature of the  
30 spray drying is preferably in the range of from 300 to 600 °C and the outlet temperature is in the range of from 130 to 190 °C, preferably of from 150 to 170 °C.

Over and above the present invention relates also to the use of said mixture of the invention or the use of said mixture prepared according to said process of the invention in the manufacture of highly pure metals, in particular tantalum, in silver  
5 soldering, as catalyst in the reaction of organic compounds, and as fluorinating agent for organic compounds, in particular for aromatic compounds.

The present invention is now described with some examples.

## 10 EXAMPLES

### Example 1 (laboratory test)

500 g water free potassium fluoride (water content < 0.3 % by weight) from a  
15 fresh production were speedily weighed out to a 2 liter polyethylene lining (low density polyethylene mono foil with a thickness of 100  $\mu\text{m}$ , 1  $\text{g}/\text{m}^2 \cdot \text{d}$ ), sealed and mixed with the indicated additives and the indicated amount of table 1. Subsequently the samples were stored in a laboratory hood at room temperature.

20 Two 500 g samples of potassium fluoride without additives (blind samples) were stored for comparison under the same conditions.

At the beginning of the experiment all samples were free from lumps and were free flowing. After six months the samples were controlled. For this purpose the  
25 samples were multiple turned and the free flowing was compared with the reference sample by visually judging. Furthermore the weight increase was determined as measure of the take-up of the water amount through the polyethylene lining. After a storage period of six months the laboratory samples containing the high dispersed silicic acid of the Aerosil® type had kept the free flowing. An insignificant  
30 formation of lumps could be detected only. Said lumps disintegrated easily using slight force. In contrast the samples containing as additives calcium phos-

phate, Tamol®, and poly(ethylene glycole), as well as the blind samples, had got hardened to a single lump. Under the influence of force the samples disintegrated to coarse pieces, not showing free flowability.

- 5 The results of the experiments are summarized in table 1:

Tab. 1

potassium fluoride spray dried	batch	consistency	weight increase g / % by weight	storage period of
+0.2 % by weight of Aerosil® 150	8229 A	few, easily crushable pieces	3.87/0.77	6 months
+0.2 % by weight of Aerosil® 380	8229 A	few, easily crushable pieces	3.78/0.75	6 months
+0.2 % by weight of Aerosil® 200	8229 A	few, easily crushable pieces	4.38/0.87	6 months
+ 1 % by weight of Aerosil® 200	8229 A	very few, easily crushable pieces	3.95/0.79	6 months
+ 0.5 % by weight of PEG 6000	8229 A	lumpy, crushable pieces	3.98/0.79	6 months
+ 0.5 % by weight of Tamol® NN 940	8229 A	joined and stucked bridges	3.96/0.79	6 months
+ 0.5 % by weight of calcium phosphate	8229 A	lumpy, bulky crushable pieces	3.77/0.75	6 months
+ blind sample	8229 A	joined and stucked bridges	3.34/0.66	6 months

- 10 Tamol® = registered trade mark of BASF, condensation product of naphthalenic sulfonic acid and formaldehyde  
PEG = poly(ethylene glycole)

15 **Example 2 (production trial)**

- An aqueous solution containing 37 % of potassium fluoride was dried in a spray dryer using hot air with an inlet temperature of 400 °C and an outlet temperature of 180 °C. By means of a differential batchweighing scale which was placed after  
20 the outlet of the spray tower pyrogenic Aerosil®200 was metered, and was fed to the potassium fluoride on the way to the cooling down via a conveying screw and

- 7 -

- mixed with said fluoride in a mixer in a manner to result in a mixture with a silicon dioxide content of 0.5 % by weight. 25 kg of the obtained mixture were stored for 6 months in a fiber drum sealed with a polyethylene lining. After the storage period of 6 months said material having been treated with the pyrogenic silicic acid resulted in a take-up of water of approximately 1 % by weight and a good free flowing. The comparison sample without said silicic acid as additive, which was stored under the same conditions had got hardened. Under influence of force the product disintegrated to coarse lumps.
- 10 A sieve analysis of the products pointed out that the lumps of the comparison material could not disintegrate completely during sieving. For the material without silicic acid the sieving residue on a 75 µm woven-wire increased from 11 % at the beginning of the storage period to 23 % at the end of the storage period. For the material treated with the Aerosil<sup>®</sup>, the sieving did not result in a significant
- 15 change after the storage period of six months. The results of the sieve analysis are summarized in table 2:

Tab. 2

potassium fluoride	sieve analysis/ consistency	ignition loss	storage period of:
+0.5 % by weight of Aerosil <sup>®</sup> 200	98 % < 150 µm, 89 % < 75 µm, free flowing	0.13 %	direct after manufacture
+0.2 % by weight of Aerosil <sup>®</sup> 200	98 % < 150 µm, 87 % < 75 µm, free flowing	1.3 %	6 months
blind sample without Aerosil <sup>®</sup>	98 % < 150 µm, 77 % < 75 µm, lumpy	1 %	6 months

- 20 Sieving with sieving machine of company Fritsch, Analysette, amplitude 1.5, sieving period 20 minutes

PCT/EP00/07950  
Honeywell Specialty Chemicals Seelze  
GmbH

11. Februar 2002  
R29699PC RI/Ben

### Claims

- 5 1. Mixture, comprising an alkali metal fluoride or an earth alkali metal fluoride or a mixture of two or more thereof and 2 % by weight or less of silicic acid, preferably pyrogenic acid.
2. Mixture as claimed in claim 1, comprising as alkali metal fluoride potassium fluoride.
- 10 3. Mixture as claimed in claim 1 or 2, characterized in that it contains of from 0.2 to 2 % by weight of silicic acid, relative to the total weight of the mixture.
- 15 4. Mixture as claimed in any of the claims 1 to 4, characterized in that it contains of from 99 to 99.8 % by weight of potassium fluoride and of from 0.2 to 1 % by weight of silicic acid, each relative to the total weight of the mixture.
- 20 5. Process for the manufacture of a mixture as claimed in any of the claims 1 to 4, characterized in that dried alkali metal fluoride or dried earth alkali metal fluoride or a mixture of two or more thereof is mixed with 2 % by weight or less of silicic acid, relative to the total weight of the mixture.
- 25 6. Process as claimed in claim 5, characterized in that the dried alkali metal fluoride or the dried earth alkali metal fluoride or the dried mixture of two or more thereof is obtained by spray drying of a solution, comprising the

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alkali metal fluoride or the earth alkali metal fluoride or the mixture of two or more thereof.

- 5        7.     Process as claimed in claim 6, characterized in that the spray drying is carried out at an inlet temperature in the range of from 300 to 600 °C and an outlet temperature of from 130 to 190 °C.
- 10      8.     Use of a mixture as claimed in any of the claims 1 to 4 or a mixture, preparable in a process as claimed in any of the claims 5 to 7, in the production of highly pure metals, in silver soldering, as catalyst in the reaction of organic compounds and as fluorinating agent for organic compounds.



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PCT/EP00/07950  
Honeywell Specialty Chemicals Seelze  
GmbH

11. Februar 2002  
R29699PC RI/Ben

### Abstract

- 5 Mixture, comprising an alkali metal fluoride or an earth alkali metal fluoride or a mixture of two or more thereof and 2 % by weight or less silicic acid, preferably pyrogenic silicic acid, a process for the production thereof and the use in the production of highly pure metals, in silver soldering, as catalyst in the reaction of organic compounds and as a fluorinating agent for organic compounds.

10

Atty. Dkt. No. 077251-0104

**DECLARATION AND POWER OF ATTORNEY**

As a below named inventor, I HEREBY DECLARE:

THAT my residence, post office address, and citizenship are as stated below next to my name;

THAT I believe I am the original, first, and sole inventor (if only one inventor is named below) or an original, first, and joint inventor (if plural inventors are named below or in an attached Declaration) of the subject matter which is claimed and for which a patent is sought on the invention entitled

FLOWABLE ALKALINE FLUORIDES AND ALKALINE EARTHS FLUORIDES(Attorney Docket No. 077251-0104)

the specification of which (check one)

     is attached hereto.

  X   was filed on August 16, 2000 as United States Application Number or PCT International Application Number PCT/EP00/07950 and was amended on          (if applicable).

THAT I do not know and do not believe that the same invention was ever known or used by others in the United States of America, or was patented or described in any printed publication in any country, before I (we) invented it;

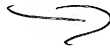
THAT I do not know and do not believe that the same invention was patented or described in any printed publication in any country, or in public use or on sale in the United States of America, for more than one year prior to the filing date of this United States application;

THAT I do not know and do not believe that the same invention was first patented or made the subject of an inventor's certificate that issued in any country foreign to the United States of America before the filing date of this United States application if the foreign application was filed by me (us), or by my (our) legal representatives or assigns, more than twelve months (six months for design patents) prior to the filing date of this United States application;

THAT I have reviewed and understand the contents of the above-identified specification, including the claim(s), as amended by any amendment specifically referred to above;

THAT I believe that the above-identified specification contains a written description of the invention, and of the manner and process of making and using it, in such full, clear, concise, and exact terms as to enable any person skilled in the art to which it pertains, or with which it is most nearly connected, to make and use the invention, and sets forth the best mode contemplated by me of carrying out the invention; and

THAT I acknowledge the duty to disclose to the U.S. Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.



Atty. Dkt. No. 077251-0104

I HEREBY CLAIM foreign priority benefits under Title 35, United States Code § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below any foreign application for patent or inventor's certificate or of any PCT international application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number	Country	Foreign Filing Date	Priority Claimed?	Certified Copy Attached?
199 39 353.2	Germany	08/19/1999	YES	

I HEREBY CLAIM the benefit under Title 35, United States Code § 119(e) of any United States provisional application(s) listed below.

U.S. Provisional Application Number	Filing Date

I HEREBY CLAIM the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application Number	PCT Parent Application Number	Parent Filing Date	Parent Patent Number

I HEREBY APPOINT the registered attorneys and agents at Customer Number 22428



22428

PATENT TRADEMARK OFFICE

to have full power to prosecute this application and any continuations, divisions, reissues, and reexaminations thereof, to receive the patent and to transact all business in the United States Patent and Trademark Office connected therewith.